

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

REMARKS

Review and reconsideration on the merits are requested.

Applicants appreciate the Examiner's **Response to Amendment**.

Applicants now address the objection to the **Specification** on the ground it introduces new matter.

The Examiner makes the following points:

- a) On page 6, in the paragraph beginning at line 7, the insertion of the phrase "polymer electrolyte membrane in the" constitutes new matter.
- b) On page 6, in the paragraph beginning at line 10, insertion of the phrase "polymer electrolyte membrane in the" constitutes new matter.
- c) In the paragraph bridging pages 8 and 9, the insertion of the phrase "the polymer electrolyte membrane having" constitutes new matter.
- d) On page 10, in the paragraph beginning at line 10, the insertion of the phrase "the polymer electrolyte membrane in" constitutes new matter.
- e) On page 30, in the paragraph beginning at line 9, the insertion of the phrases "the polymer electrolyte membrane in" and "of a single cell" constitutes new matter.
- f) On page 31, in the paragraph beginning at line 8, insertion of the phrase "polymer electrolyte membrane in the" constitutes new matter.

Applicants have carefully reviewed the specification and the above amendments, and respectfully must submit that no new matter has been introduced in the specification when the overall disclosure in the specification is considered.

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

Applicants believe that the following discussion will make it abundantly clear that the Q value is specific to the polymer electrolyte membrane, not to the membrane electrode assembly. Accordingly, Applicants submit that the changes they have made with the specification in no matter can be seen to reflect new matter.

Applicants address the points as presented.

Point a), Page 6, paragraph beginning at line 7

The language is as follows:

Fig. 2 is a schematic cross-sectional view showing an apparatus for measuring the Q value of the polymer electrolyte membrane in the membrane electrode assembly of the present invention;

The issue presented is whether Fig. 2 shows an apparatus for measuring the Q value of the polymer electrolyte membrane in the membrane electrode assembly.

The Examiner's attention is first directed to the specification at page 8, last line to page 9, line 2, "At least in the second membrane electrode assembly, the polymer electrolyte membrane should have a Q value (charge per a unit area) of 0.09 - 0.18 C/cm²."

The specification continues at page 9, lines 2-6 and states (*italics added*):

"When the Q value is less than 0.09 C/cm², it is impossible to obtain sufficient power-generating performance. On the other hand, when it exceeds 0.18 C/cm², the *polymer electrolyte membrane* has too low heat resistance, resulting in too high percent defective."

The Examiner is also referred to the specification at page 8, lines 4/5, where the following disclosure occurs (*italics added*) "Particularly, in the case of forming a *polymer*

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

electrolyte membrane having a softening point of 120°C or higher *and a Q value of 0.09 - 0.18 C/cm²*, it is preferable to use a sulfonated hydrocarbon polymer.”

Quite clearly it is the Q value which is specific to the polymer electrolyte membrane, not to the membrane electrode assembly, which is at issue. Thus, one reading the specification would understand that the apparatus of Fig. 2 is for measuring the Q value of the polymer electrolyte membrane, not the membrane electrode assembly.

Point b), Page 6, paragraph beginning at line 10

Fig. 3 is a graph showing a discharge curve obtained as a result of measuring a current density in a predetermined voltage range, to determine the Q value of the polymer electrolyte membrane in the membrane electrode assembly of the present invention;

Again, the Examiner is referred to the earlier mentioned disclosure at page 8, last line to page 9, line 2, page 9, lines 2-6 and page 8, lines 4/5.

Considering all of this disclosure, it is quite clear that the Q value *of the polymer electrolyte membrane* is measured and that is 0.09 - 0.18 C/cm², i.e., the Q value is specific to the polymer electrolyte membrane and not the membrane electrode assembly.

There is absolutely nothing in the specification, other than the few corrections that Applicants make, which would suggest that the Q value as defined would apply to the membrane electrode assembly.

Point c), paragraph bridging page 8/9

The Q value may be regarded as an indicator of adhesion of the electrode to the polymer electrolyte membrane, and it has been found that with the polymer electrolyte membrane having the Q value of 0.09-0.18 C/cm², an excellent polymer electrolyte membrane electrode assembly is obtained.

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

Again, Applicants refer the Examiner to the above discussion.

In fact, reviewing the change at Point c), it is only internally consistent if the phrase --the polymer electrolyte membrane having-- is included.

Point d), paragraph beginning at page 10, line 10

In the discharge curve shown in Fig. 3, the Q value is defined as the amount of electric charge per a unit area of the polymer electrolyte membrane in the membrane electrode assembly, indicating that the larger the Q value, the higher the adhesion of the electrode 100 to the polymer electrolyte membrane 101.

See especially the earlier cited material from the specification especially at page 8, last line to page 9, line 2; the Q value is specific to the polymer electrolyte membrane, not to the membrane electrode assembly.

Point e), paragraph beginning at page 30, line 9

The Q value of the polymer electrolyte membrane in each membrane electrode assembly of a single cell in EXAMPLES 8-11 and COMPARATIVE EXAMPLES 4 and 5 was measured in a range from -0.1 V to +0.7 V. The measurement results are shown in Table 2.

Again, Applicants refer the Examiner to the disclosure of page 8, last line to page 9, line 2 but, in addition, refer to page 30, lines 9-21 as well as Fig. 2 and Examples 8-11. Referring to page 9, for the reasons earlier advanced, quite clearly the disclosure at page 30, lines 9-12 would refer to Q value of each polymer electrolyte membrane which would be if in a membrane electrode assembly.

Referring now to page 30, lines 14-21, reference is quite clearly to a single cell at page 30, line 15.

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

Referring to Fig. 2, Fig. 2 shows the polymer electrolyte membrane 101 in a measuring position.

Examples 8-11 relate to the use of a single cell. It must be remembered that the polymer electrolyte membrane is *in* a membrane electrode assembly, but from Fig. 2, it can be seen that it is the polymer electrolyte membrane which is undergoing measurement, and it is the Q value of the polymer electrolyte membrane in each membrane electrode assembly of a single cell which is being tested or evaluated in Examples 8-11.

Thus, Examples 8-11 use a single cell comprising each membrane electrode assembly produced by using a polymer electrode membrane which is sandwiched by an oxygen electrode and a fuel electrode each comprising a gas-diffusion layer carrying a catalyst (or having a catalytic layer formed thereon) followed by hot pressing; see the specification at page 5, lines 7-16 and page 27, line 18 to page 28, line 26 in view of page 30, lines 14-18.

However, Applicants propose to further amend as later discussed.

Point f), paragraph beginning at page 31 line 8

As is clear from Table 2 and Fig. 11(a), when the Q value of the polymer electrolyte membrane in the membrane electrode assembly is less than 0.09 C/cm², only low voltage is generated.

Page 8, last line to page 9, line 2 clearly supports the change Applicants have made. The Q value is always defined with specificity as referring to the polymer electrode membrane, not the membrane electrode assembly.

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

All Applicants have done with all of the above changes is simply to make corrections to the specification which would be completely clear and accepted by one of ordinary skill in the art.

Withdrawal is requested.

Applicants now turn to the **objection to the disclosure because of various formalities beginning at page 3 of the present Action.**

Applicants address the Examiner's objections in the order presented.

Applicants first address the objection to paragraph 39 of the specification which would correspond to the paragraph bridging pages 8/9, which Applicants have amended.

Applicants further amend at page 9, lines 10-13 to call for "a polymer electrode membrane in the membrane electrode assembly".

This amendment finds support at page 9, lines 22-24 and Fig. 2 of the specification. At page 9, lines 22-24, it is stated (*italics added*):

"The *polymer electrolyte membrane* 101 is in contact with an aqueous sulfuric acid solution 109 of pH 1 on a free side from the electrode 100 and with a nitrogen gas on the side of the electrode 100."

Quite clearly, when read in conjunction with Fig. 2, the above disclosure supports the original Amendment to page 9, lines 10-13.

Applicants now address the Examiner's position regarding what the value Q means in Paragraph 39, which would appear to correspond to the paragraph bridging pages 8/9 of the specification, the Examiner specifically indicating that it appears to be a contradiction of what the Q value is since the specification states that the polymer electrolyte membrane should have a

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

certain Q value and at the same time the specification states that the Q value may be regarded as an indicator of adhesion of the electrode to the polymer electrolyte membrane.

Though Applicants see no inconsistency in the paragraph bridging pages 8/9 of the specification at the above points, Applicants respond with the following remarks.

From the above current amendment to the paragraph bridging pages 8/9 of the specification, Applicants submit it is abundantly clear that the Q value is the amount of electric charge per a unit area determined from a peak area of proton on an adsorption side in the scanning of voltage from -0.1 V to +0.7 V, in a cell in which the amount of platinum in the catalytic layer of each electrode is 0.5 mg/cm², where the polymer electrode membrane in the membrane electrode assembly is surrounded by an aqueous sulfuric acid solution of pH 1 on one side and a nitrogen gas on the other side, wherein Q can be determined from *the proton peak area* in the discharge curve on the adsorption side with a scanning voltage from -0.1 V to +0.7 V, as shown in Fig. 3 and as mentioned at page 10, lines 10-12 of the specification.

In more detail, because the Q value is defined as the amount of electric charge per a unit area of the polymer electrode membrane in the membrane electrode assembly, the proton peak area in the discharge curve on the adsorption side corresponds to the total amount of electric charge adsorbed in the cell with a scanning voltage from -0.1 V to +0.7 V, the cell having a predetermined and constant area of the electrode, whereby the larger the Q value, the higher the adhesion of the electrode 100 to the polymer electrolyte membrane 101, since the total amount of electric charge depends on resistance between the polymer electrolyte membrane and the electrode adhered thereon (see page 10, lines 13-16 of the Specification).

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

Accordingly, it is believed quite accurate and not inconsistent with any other portion of the specification to say that:

“The Q value may be regarded as an indicator of adhesion of the electrode 100 to the polymer electrolyte membrane 101.”

Certainly the Examiner has advanced no reason as to why statements regarding the catalyst loading and adhesion are inconsistent.

The Examiner next refers to Paragraph 42, which again relates to adhesion, and it is believed that the above remarks respond to the Examiner’s position on Paragraph 42.

Turning finally to the Examiner’s statement regarding Paragraph 135, which would correspond with the paragraph beginning at page 30, line 9, Applicants propose amend as follows on page 30 and page 35:

Proposed Amendment

They propose to amend “the Q value of each membrane electrode assembly in EXAMPLES 8-11 (or 12-16) and COMPARATIVE EXAMPLES 4 and 5 (or 6 and 7) was measured in a range from -0.1 V to +0.7 V.” at page 30, lines 9-11 and page 35, lines 3-5 of the specification to --the Q value of each membrane electrode assembly comprising polymer electrolyte membrane in EXAMPLES 8-11 (or 12-16) and COMPARATIVE EXAMPLES 4 and 5 (or 6 and 7) was measured in a range from -0.1 V to +0.7 V--, respectively.

These amendments find support at page 7, lines 20-23 and page 31, lines 5-6 (Table 2) of the specification.

Applicants would like to discuss the above amendment in a telephone interview with the Examiner.

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

Applicants believe that the above responds to all objections to the specification, and request withdrawal.

Applicants now address the rejection of claims 5-9, 16 and 17 under 35 U.S.C. § 112, second paragraph, which begins at the top of page 5 of the Action.

Claim 5 of the present application calls for that: "A membrane electrode assembly comprising a polymer electrolyte membrane, said polymer electrolyte membrane having a softening point of 120°C or more and a Q value of 0.09-0.18 c/cm²."

One feature of claim 5 is that the polymer electrolyte membrane has a Q value of 0.09-0.18 C/cm².

As is clear from the above discussions on the Q value regarding the objections to the specification, the Q value is defined as the amount of electric charge per a unit area of the polymer electrode member in the membrane electrode assembly, the proton peak in the discharge curve on the adsorption side corresponding to the total amount of electric charge adsorbed with a cell by a scanning voltage from -0.1 V to +0.7 V, where the cell has a predetermined and constant electrode area (see e.g., the specification at page 30, lines 8-12; page 31, lines 8-14 and Fig. 11(a) (Examples 8-11), and Table 2 at page 31 of the specification.

Accordingly, the Q value is specific to a polymer electrolyte membrane, and the greater the Q value, the higher the adhesion of the electrode 100 to the polymer electrolyte membrane 101, since the total amount of electric charge depends on resistance between the polymer electrolyte membrane and the electrode adhered thereon (see page 10, lines 13-16 of the

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

specification), so that the Q value serves to indicate the degree of adhesion of the electrode on the polymer electrolyte membrane.

The prior art: U.S. 6,136,412 Spiewak et al (Spiewak); U.S. 5,273,017 Swathirajan et al (Swathirajan); U.S. 5,795,496 Yen et al (Yen).

The Prior Art Rejections

Applicants first address the anticipation rejection of claims 1, 4 and 12 over Spiewak. The Examiner cites certain portions of Spiewak to support the rejection.

Applicants traverse as follows.

Claim 1 of the present application reads as follows:

1. A membrane electrode assembly comprising a pair of opposing electrodes each having a catalytic layer, and a polymer electrolyte membrane sandwiched by said electrodes, part of said catalytic layers being projecting into said polymer electrolyte membrane, wherein the projection depth of said catalytic layer is 0.5 μ m or more and less than 5 μ m.

As is shown in Fig. 5 of the present application, when the average projection depth D is less than 0.5 μ m, sufficient contact cannot be made between the catalytic layer and the polymer electrolyte membrane. This results in insufficient cross-leaking, which will, in consequence, result in insufficient self-humidifying.

In distinction, when the average projection depth D is 5 μ m or more, excessive cross-leaking takes place. A more preferred average projection depth D is 0.5 - 3 μ m. See the specification at page 12, lines 21-27.

Spiewak is quite in contrast with claim 1 of the present application.

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

Spiewak discloses a membrane electrode or membrane electrode assembly (MEA) comprising an ion conducting membrane (ICM) and one or more electrode layers that comprise nanostructured elements. The nanostructured elements further comprise a catalytic material, and the nanostructured elements are in complete contact with the ICM so that greater than 0% and less than 99% of the volume of each of such elements is embedded in the ICM, more specifically, in the membrane electrode assembly (MEA) catalyst electrodes are incorporated into very thin surface layers, localized within 2 μm of the interface on the outer side of an ion conductive membrane (ICM). See Spiewak in column 3, lines 17-24, column 7, lines 21-25, column 8, line 10 and Figs. 2 and 3. Spiewak is completely silent with regarding the projection depth of the catalytic layer.

In more detail, Figs. 5(a), 5(b) and 5(c) of Spiewak are scanning electron micrographs of a cross section of an MEA surface where the nanostructured electrode layer conforms to a microstructure shape of 25 μm peaks and valleys (the projection depth of the catalytic layer) as obtained from Figs. 5(a) and 5(b), taken at 500x (60.0 μm in unit-length), 5,000x (1,5000x shown in Fig. 5(b)) (20.0 μm in unit-length), respectively, and Figs. 5(a) and 5(b) show that the actual electrode layer surface area per unit planer area of the MEA is increased by the geometric surface area factor of the microtextured substrate, i.e., at a factor of 1.414 or the square root of two (2), since each part of the surface is at a 45° angle to the normal stacking axis (see column 20, lines 53-64).

Thus, Spiewak does not teach or suggest that the projection depth of any catalytic layer is 0.5 μm or more and less than 5 μm .

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

Accordingly, since the Examiner has offered no reasonable ground to conclude that the projection of any catalytic layer in Spiewak would meet that of claim 1 herein, Applicants respectfully submit that the anticipation rejection is improper and should be withdrawn.

Applicants rely on their arguments regarding claim 1 for the patentability of claims 4 and 12.

Withdrawal is requested.

Applicants now address the rejection of claims 1, 4 and 12 as anticipated by Swathirajan.

Applicants traverse as follows.

Swathirajan may disclose a method where a catalyst slurry is heated while being pressed to a membrane for a time/temperature/compressive load sufficient to soften the membrane and at least partially embed at least a portion of the catalyst particles in the membrane (see column 2, lines 22-26). However, Swathirajan fails to teach or suggest any projection of the catalyst layer as in claim 1 of the present application.

This reason for this statement is that in Swathirajan the step of heating while pressing is conducted under a high pressure of at about 1,000 to about 2,000 pounds per square inch (1b/in²) (6.9-13.7 MPa) - a compressed load - for about 1 to 5 minutes at a temperature of about 120°C to about 150°C (see column 2, lines 5-155). This is quite contrary to the conditions of the present invention which are under a lower pressure than Swathirajan and involve one step at a temperature of 60-200°C and a pressure of 1-10 MPa for 1-5 minutes (Examples 8-13 and 16: 2.5-3.0 MPa and 120-160°C for 1-2 minutes) or two steps at a first hot-pressing at a relatively low temperature, and then a second hot-pressing at a relatively higher temperature for a short

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

period of time such that the first hot pressing conditions are at about 60-100°C (for instance, about 80°C) and about 1-10 MPa (for instance, about 2.5 MPa) for about 1-5 minutes (for instance, 2 minutes) and a second hot pressing at about 120-200°C (for instance, 160°C) and about 1-10 MPa (for instance, about 3 MPa) for about 1-5 minutes (for instance, 1 minute) (see page 18, lines 5-14 of the specification).

Since there is no basis, even inherency, to support the Examiner's anticipation rejection, Applicants request withdrawal.

They rely upon their arguments for claim 1 above with respect to claims 4 and 12.

Withdrawal of the rejection is requested.

Applicants now address the rejection of claim 10 as obvious over Spiewak in view of Yen.

Claim 10 is currently amended calls for:

10. (currently amended): A membrane electrode assembly comprising a pair of opposing electrodes each having a catalytic layer, and a polymer electrolyte membrane sandwiched by said electrodes, part of said catalytic layers being projecting into said polymer electrolyte membrane, wherein the projection depth of said catalytic layer is 0.5 μ m or more and less than 5 μ m, and

wherein said polymer electrolyte membrane is made of a sulfonated hydrocarbon polymer that may contain oxygen in its skeleton or other substituent groups than a sulfonic group, wherein the sulfonated hydrocarbon polymer is selected from the group consisting of sulfonated polyetheretherketone, sulfonated polysulfone, sulfonated polyethersulfone, sulfonated polyetherimide, sulfonated polyphenylene sulfide and sulfonated polyphenylene oxide.

Considering claim 10, one major distinguishing feature of the present invention lies in the fact that the projection depth of the catalytic layer of each electrode constituting a pair of opposing electrodes of the membrane electrode assembly is 0.5 μ m or more and less than 5 μ m.

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

As a consequence, the membrane electrode assembly will exhibit an excellent self-humidifying function so that low-humidifying operation can be carried out without sacrificing power-generating performance in a polymer electrolyte fuel cell formed by stacking such membrane electrode assemblies via separator plates. See page 37, lines 4-11 of the present specification.

A further distinguishing feature lies in the fact that the polymer electrolyte membrane is made of a sulfonated hydrocarbon polymer that may contain oxygen in its skeleton or other substituent groups than a sulfonic group, the sulfonated hydrocarbon polymer being selected from the group consisting of sulfonated polyetheretherketone, sulfonated polysulfone, sulfonated polyethersulfone, sulfonated polyetherimide, sulfonated polyphenylene sulfide and sulfonated polyphenylene oxide (page 4, line 28 to page 5, line 6 of the specification), whereby one can form a polymer electrolyte membrane having a softening point of 120°C or higher and a Q value of 0.09-0.18 C/cm². To achieve these results, it is necessary for the sulfonated hydrocarbon polymer as such to have a preferred ion exchange capacity (milli-equivalent of the sulfonic group per 1 g) of 1-2.6 meq/g. See the specification page 8, lines 4-6 and lines 16-17.

Both of the above factors are important factors in claim 10.

From the earlier discussion regarding Spiewak, it is clear that Spiewak fails to teach or suggest the former distinguishing feature of claim 10 above discussed and, with respect to the latter distinguishing feature, although Spiewak may teach the use of a sulfonic acid-type resin or other polymers which exhibit cation-exchange capability upon sulfonation, and mention is made of "NAFION", Spiewak is silent regarding the necessary properties required for the sulfonated hydrocarbon polymer to preferably have an ion exchange capacity (milli-equivalent of the

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

sulfonic group per 1 g) of 1 - 2.6 meq/g, which permits one to obtain a polymer electrolyte membrane having a Q value of 0.09 - 0.18 C/cm².

Turning now to Yen, Yen discloses truly sulfonated polyetheretherketone stable at a higher temperature of 200-300°C as an electrolyte membrane for fuel cells compared with "NAFION" (see column 1, line 66 to column 2, line 3). However, Yen fails to teach or suggest either of the features of the amended claim 10 of the present application discussed above.

Accordingly, Applicants respectfully submit that one of ordinary skill in the art, referring to the combination of Spiewak and Yen, would not be led to the invention of claim 10 and, accordingly, claim 10 is not obvious over Spiewak in view of Yen.

Applicants now address the rejection of claim 10 as obvious over Swathirajan in view of Yen.

They traverse as follows.

At the bottom of page 8 of the Action, the Examiner states:

"Swathirajan et al (US Patent 5,272,017) disclose all the limitations of claim 10 above except that the sulfonated hydrocarbon polymer is sulfonated polyetheretherketone. Swathirajan et al do disclose NAFION as the ion exchange resin in the membrane (col. 4, lines 42-50)."

However, as has been earlier discussed, amended claim 1 is not anticipated by Swathirajan because Swathirajan fails to teach or suggest any projection of the catalyst layers as recited in claim 1, whereby one following the present invention can obtain an excellent self-humidifying function which permits carrying out low-humidifying operation without sacrificing power generating performance in a polymer electrolyte fuel cell formed by stacking the membrane electrode assembly via separator plates.

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

Focusing on sulfonated hydrocarbon polymers, Swathirajan is silent on this point except for mentioning commercially available sulfonated perfluorocarbons such as “NAFION,” and the like (as indicated by the Examiner). However, “NAFION” does not have a sulfone (-SO₂-) group in its polymer main chain (or polymer backbone), rather, it has a plurality of terminal groups of the formula -O-CF₂CF(CF₃)-OCF₂CF₂-SO₃Na branching from the polymer main chain of “NAFION”, which is different from a sulfonated hydrocarbon polymer such as a sulfonated polysulfone polymer constituting the polymer electrolyte membrane used in the present invention (see column 4, lines 42-57).

Quite clearly Swathirajan alone does not render claim 10 obvious.

However, the rejection is a combination rejection, and Applicants now turn to Yen.

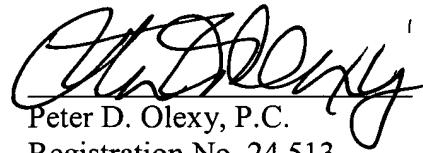
With respect to Yen, although the Yen sulfonated polyetheretherketone is stable at a higher temperature of 200-300°C as an electrolytic membrane for fuel cells as compared with “NAFION” (see column 1, line 66 to column 2, line 3), Yen fails to teach or suggest any of the distinguishing features of claim 10 of the present application as above discussed.

AMENDMENT UNDER 37 C.F.R. § 1.116
U.S. Application No.: 10/050,518

Accordingly, Applicants respectfully submit that the combination of Swathirajan and Yen does not render claim 10 obvious and, accordingly, they request withdrawal of the rejection.

Allowance is requested.

Respectfully submitted,



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